

## Response to Reviewers

We thank the reviewers for their constructive comments and suggestions, and the editor for overseeing the process. We have attempted to address all the reviewers' comments, which we feel has improved the manuscript. The major change involves expanding the section on mixtures, which both reviewers requested; in particular, we now include results from a mixture of less-similar species (a monoterpene and a ketone). We also incorporate a number of references that the reviewers suggest, providing more context to the work. Reviewer comments and our point-by-point responses are included below.

### Reviewer 1

*This manuscript provides an interesting exploration of the varying sensitivities of different VOC sensors in an array. The plots are very clear and useful for considering how a multi-sensing-technology (PID, MOx, EC) array can be used to gather species (or species-group) specific information. However, this is not the first time sensor arrays have been used to measure VOCs, yet the manuscript does not seem to fully acknowledge much of the other work on VOC sensing and sensor arrays, making it difficult to fully assess the impact of this work. A number of suggestions are given below to better contextualize this work within the related body of VOC sensing efforts.*

Response: We thank the reviewer for the thorough and constructive evaluation of our manuscript. We are particularly grateful for highlighting key papers in the literature that were not cited in our original submission. As described below, we now include these additional citations that provide more context for the present work.

*Line 78: More recent examples of e-noses should be given here, as there have been many developments in this space since 1994.*

Response: The manuscript has been edited to include more recent examples (Lotesoriere et al., 2024; Vadera and Dhanekar, 2025).

*Line 83: You mention that sensor sensitivities to non-VOC gases are a challenge for VOC sensors. Why was this not explored in the present study, alongside the exploration of RH and VOC mixture impacts? If it cannot be added into the study, the reasoning for excluding it, as well as suggested avenues for testing these impacts, should at least be mentioned.*

Response: It's true that some commercially-available VOC sensors have cross-sensitivities to non-VOC gases. PIDs generally do not have such cross-sensitivities, but some EC and MOx sensors are sensitive to gases such as CO, CO<sub>2</sub>, CH<sub>4</sub>, H<sub>2</sub>, NO<sub>2</sub>, H<sub>2</sub>S, or SO<sub>2</sub> (Spinelle et al., 2017). We deliberately chose EC and MOx sensors without known cross-sensitivities for inclusion in our sensor array. We have edited the manuscript to include this reasoning for sensor selection and lack of testing for impacts of non-VOC gases.

*Paragraph starting on line 85: Examples of prior MOx sensor arrays are given. Examples of successful uses of PID sensors should also be mentioned (even if they are not arrays) to give context to the prior use of these sensors.*

Response: The manuscript has been edited to include studies that use PID sensors for measuring VOCs (Ding et al., 2023; Yang et al., 2026)

*Line 97-98: There actually are examples using a range of sensing technologies that should be acknowledged here. Two examples: Xu, Linjie, et al. "Hybrid gas sensor array to identify and quantify low-concentration VOCs mixtures commonly found in chemical industrial parks." *IEEE Sensors Journal* 22.13 (2022): 13434-13441. And Fumian, Francesca, et al. "c." *The European Physical Journal Plus* 136.9 (2021): 913.*

*There are also many examples of using sensor arrays with temperature cycling that are worth mentioning in the introduction, as they have provided another route to gain species-specific information using VOC sensors. One such example: Baur, Tobias, et al. "Field study of metal oxide semiconductor gas sensors in temperature cycled operation for selective VOC monitoring in indoor air." Atmosphere 12.5 (2021): 647.*

Response: We thank the reviewer for suggesting these examples from the literature. We have edited the manuscript to reference this prior work and explicitly acknowledge that our array is not the first to take this approach. Specifically, we have included the following text:

“While most of these arrays use only MOx sensors, some recent studies have leveraged multiple sensing technologies to measure target compounds. For example, Xu et al., 2022 used an array of four different metal oxide sensors, one photo-ionization detector, and one electrochemical sensor to measure elevated levels (0.5-5 ppm) of three VOCs (toluene, dichloromethane, and ethyl acetate). Similarly, Silberstein et al. (2024) used an electrochemical sensor in addition to several MOx sensors to quantify methane emissions from an oil well. These studies demonstrate the potential of air sensor arrays to generate quantitative, chemically specific VOC information.”

*Lines 125-126: This is not true. Temperature control has been used many times in VOC sensing. Mostly in indoor applications, but since this work is in a controlled laboratory setting, it cannot exclude indoor applications in its analysis: See above with work by Baur et. al., as well as He, Junjie, et al. "Low-cost MOX sensor for indoor ppb-level VOC detection using pulsed temperature-voltage dual modulation." Sensors and Actuators B: Chemical (2025): 138289. And Leidinger, M., et al. "Selective detection of hazardous VOCs for indoor air quality applications using a virtual gas sensor array." Journal of Sensors and Sensor systems 3.2 (2014): 253-263. And On the performance evaluation of hybrid and mono-class sensor arrays in selective detection of VOCs: A comparative study*

Response: We again thank the reviewer for these examples from the literature. We have edited the manuscript to reference this prior work and explicitly acknowledge that our array is not the first to include temperature control of metal oxide sensors. Specifically, we have included the following text:

“In fact, temperature control has been shown to increase both MOx array sensitivity (He et al., 2025) and selectivity (Srivastava and Dravid, 2006) to VOCs, including common indoor VOCs in the < 1 ppm range (Baur et al., 2021; Leidinger et al., 2014).”

*Lines 138-140: This is also not true. VOC EC sensors have been used for methane detection (Silberstein, Jonathan, Matthew Wellbrook, and Michael Hannigan. "Utilization of a Low-Cost Sensor Array for Mobile Methane Monitoring." Sensors 24.2 (2024): 519.) and for VOCs: Mayer, Thomas, et al. "Toward an Event-Based and Quality-Assured Air Sampling: A Portable System for Sensing and Sampling Volatile Organic Compounds." Analytical Chemistry 97.43 (2025): 23765-23772.*

Response: We again thank the reviewer for these examples from the literature and have edited the manuscript to reference this prior work.

*Line 138-140: Although VOC EC sensors have indeed been used before (see above), they certainly are less common. Is there any explanation for why this is that can be added?*

Response: This is due to more recent advancements in technology—specifically the development of Alphasense’s VOC-B1 sensor which has much higher sensitivity than their previous offering (ETO-B1). For example, both studies

suggested in the previous comment used this newer Alphasense EC in their arrays. We have edited the manuscript to briefly explain this effect. Specifically, we have included the following text:

“EC sensors that measure VOCs non-specifically have commonly been marketed for personal protection and industrial hygiene applications, but recent advancements in technology have allowed for usage in some atmospheric and air quality contexts (Mayer et al., 2025; Silberstein et al., 2024).”

*Lines 232-233: Why were these compounds chosen? Also, what are the typical ambient concentrations of these compounds? Is 5 ppb a reasonably low detection limit for all of these? It seems like some would often be much lower.*

Response: These compounds were chosen as a broadly representative set of atmospheric VOCs: these compounds are both common in the atmosphere (Goldstein and Galbally, 2007) and have diverse physicochemical properties. We also chose these compounds in particular because of their inclusion in an indoor air chemistry measurement campaign (Farmer et al., 2025) where our sensor array made measurements. We have added the following text:

“This list, based on the “chemical cocktail” used in a recent indoor chemistry experimental intensive (Farmer et al., 2025), includes VOCs from both natural and anthropogenic sources, and that contain several different functional groups.”

It is true ambient concentrations of these compounds are typically lower than 5 ppb, but the tested concentration range is low enough to be relevant for ambient conditions where VOC concentrations are typically in the tens (or even 100s) of ppb, such as indoor air (Brown et al., 1994) and wildfire smoke (Liang et al., 2022). We have added the following text:

“While these mixing ratios are higher than typical outdoor ambient levels, they are relevant to environments such as indoor air (Brown et al., 1994) and wildfire smoke (Liang et al., 2022) where VOC levels can exceed 10s of ppb.”

*Figure 4 (and related discussion): This figure is really interesting, as it shows the different chemical “fingerprints” produced using the sensor array. However, these fingerprints are only clear when there is only one compound present. What happens in ambient air when there is a complex mixture of compounds? Prior work has used pattern recognition / machine learning to disentangle these effects. Some discussion of how the information provided by the array could actually be used in an environmental application is needed. The following reference provides some ideas for interpretation: Rath, Ronil J., et al. "Chemiresistive sensor arrays for gas/volatile organic compounds monitoring: a review." *Advanced Engineering Materials* 25.3 (2023): 2200830.*

Response: We thank the reviewer for these suggestions and have edited the manuscript to include some possible avenues for interpreting the “fingerprints.” Specifically, we have specified the following:

“Such “fingerprints” could be good candidates for pattern recognition techniques such as linear discriminant analysis and support vector machines that have previously been used to classify and quantify VOCs (Rath et al., 2023).”

*Line 302: Why were these two compounds chosen for the RH testing? Wouldn't it have been more useful to choose two more chemically different species (i.e. an alkene and an aromatic) to understand how the effects vary more broadly? This choice should be justified.*

Response: It's true that our chosen compounds ( $\alpha$ -pinene and isoprene) are both alkenes, but we believe our results justify this intentional choice. In the manuscript, we show that the sensor array's responses to these two compounds in the same class vary drastically, highlighting the unpredictable effects of RH even for chemically similar compounds. We have added the following to the manuscript:

“Because isoprene and  $\alpha$ -pinene are both alkenes, our results also highlight the unpredictable effects of RH even for chemically similar compounds. We would expect two more chemically different species to have even larger differences in RH response and pose similar measurement problems.”

*Line 350: Again, why were two chemically-similar alkenes chosen for the binary mixture? Would the additive nature of the sensors also apply in a mixture that contains an alkene and an aromatic? It seems as if the response would be very different, which limits the applicability of these findings.*

Response: Unlike the previous comment on using similar compounds for RH testing, we agree with the reviewer that using two similar components for the binary mixture limits the applicability of our findings. We have edited the manuscript to include sensor responses to a binary mixture using two chemically dissimilar compounds:  $\alpha$ -pinene (a monoterpene) and 2-heptanone (a ketone). This includes edits to figures 7 and 8 (and additional information in the SI) that include results of this second mixture. To summarize our results, PID sensors show linear additivity. EC sensors are unable to detect 2-heptanone beyond the signal-to-noise threshold specified in the manuscript, and thus we are unable to infer their behavior to the mixture. MOx sensor responses again demonstrate that linear additivity is a bad fit, and that the addition of an interaction term more closely approximates sensor responses. Importantly, these results support our original conclusion that lab-based characterization is not a feasible approach for characterizing VOC sensor arrays, instead requiring in-field testing.

Specifically, we have edited Section 3.3 in the text to the following, with changes highlighted in orange:

Real-world environmental VOCs are almost always present within mixtures, which poses a measurement problem for non-specific VOC sensors such as the ones examined here. Additivity of air sensor responses to mixture components would help to simplify this problem, but because of the potential for cross-interferences between VOCs such additivity is not a given. **To investigate this, we exposed the sensor array to two different binary mixtures, each in two different ratios: 1-hexene and 1-octene (molar ratios of 1:1 and 3:1, representing very similar compounds), and  $\alpha$ -pinene and 2-heptanone (molar ratios of 1:10 and 3:1, representing quite different compounds).**

Figure 7 shows responses of the high-sensitivity PID to the 1-hexene/1-octene **and  $\alpha$ -pinene/2-heptanone mixtures.** (Figures S2 and S3 show the responses for additional PID and EC sensors, and Table S2 in the Supporting Information summarizes the fitted sensitivities to each mixture.) For the high-sensitivity PID, the mixture measurement is consistent with linear additivity: sensitivity is  $2.5 \times 10^{-1} (\pm 4.2 \times 10^{-3})$  mV/ppb to 1-hexene and  $1.2 \times 10^{-1} (\pm 2.1 \times 10^{-3})$  mV/ppb to 1-octene, and the observed sensitivity to the 1:1 mixture is  $1.7 \times 10^{-1} \pm (2.3 \times 10^{-3})$  mV/ppb (expected  $1.8 \times 10^{-1}$  mV/ppb), while the observed sensitivity to the 3:1 mixture is  $2.0 \times 10^{-1} (\pm 3.4 \times 10^{-3})$  mV/ppb (expected  $2.1 \times 10^{-1}$  mV/ppb). The 10.0 eV PID (Figure S2) also showed a proportional response to the 1:1 mixture, and the observed 3:1 sensitivity falls close to the expected value. **Similarly, for the  $\alpha$ -pinene and 2-heptanone mixture, the PID sensitivity is  $3.5 \times 10^{-3} (\pm 2.4 \times 10^{-4})$  mV/ppb to  $\alpha$ -pinene and  $1.6 \times 10^{-3} (\pm 2.6 \times 10^{-4})$  mV/ppb to 2-heptanone, and the observed sensitivity to the 3:1 mixture is  $2.7 \times 10^{-1} (\pm 1.2 \times 10^{-2})$  (expected  $2.9 \times 10^{-1}$  mV/ppb).** The EC sensor signals (Figures S2 and S3) also obey additivity within uncertainty, though the high measurement noise makes the EC mixture response uncertainties much higher than those of PIDs. The observed linearly additive nature of these sensor responses to mixtures is consistent with expectations based on the principles of operation of PID and EC sensors (Baron and Saffell, 2017; Freedman, 1980).

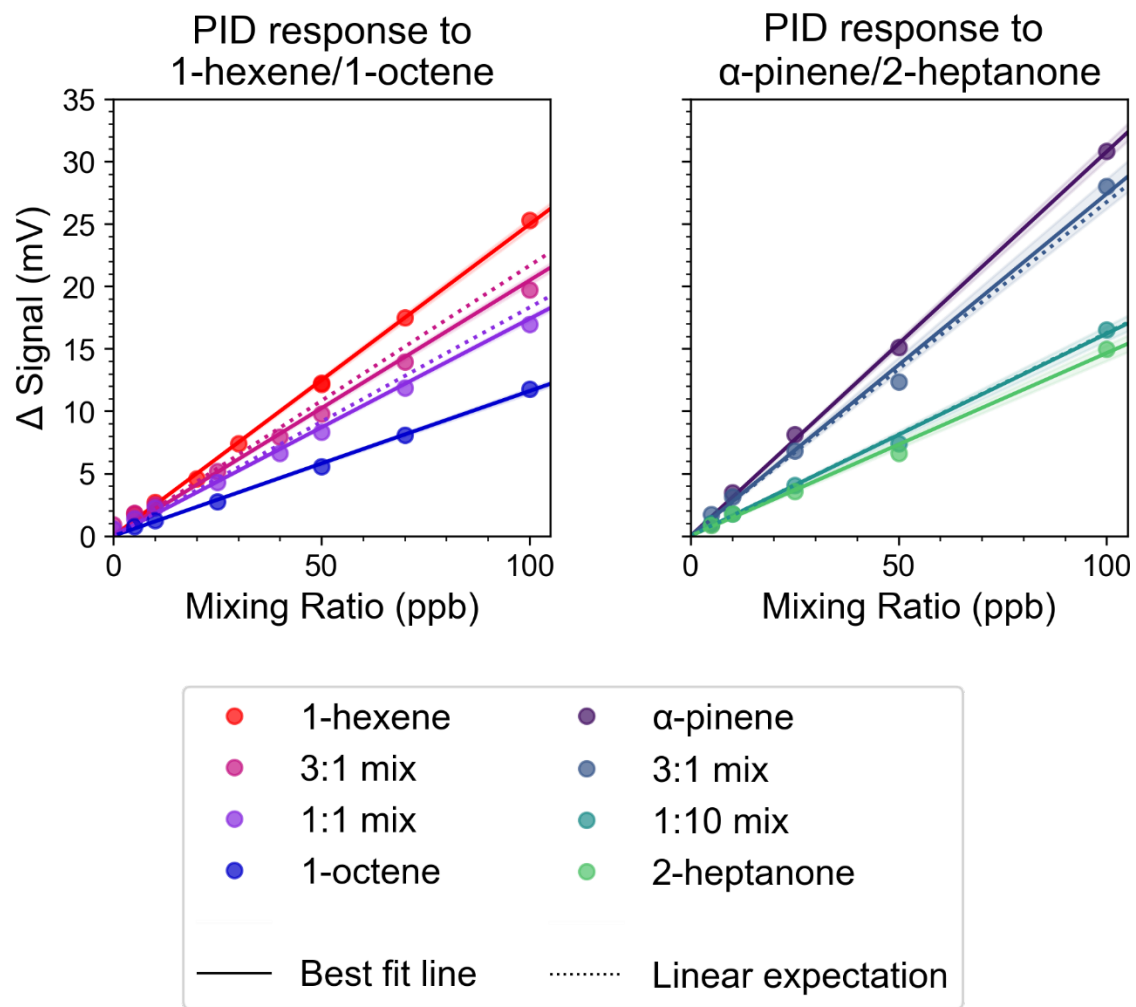
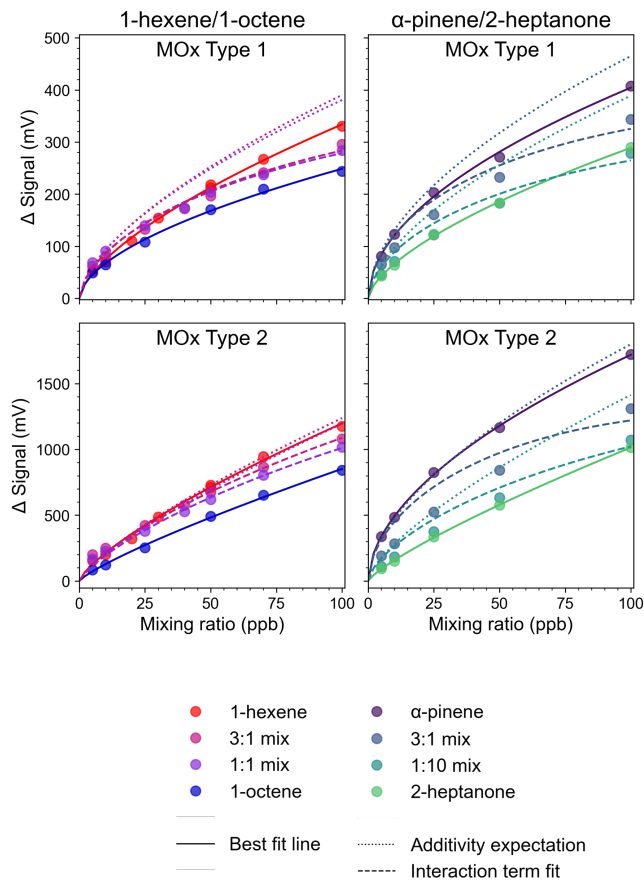


Figure 7: Left panel: high-sensitivity PID (PID2) responses to 1:1 (blue) and 3:1 (dark purple) mixtures of 1-hexene (red) and 1-octene (magenta). Right panel: PID2 responses to 3:1 (blue) and 1:10 (teal) mixtures of  $\alpha$ -pinene (indigo) and 2-heptanone (aquamarine). Solid lines indicate the best fit line for each of the points, and the shaded area indicates the  $1\sigma$  confidence interval; dotted lines indicate the expected signal assuming additivity. The mixing ratio (x axis) refers to the total amount of VOC (i.e., both components of the mixtures) introduced to the sensors.

The response of MOx sensors to binary mixtures is more complex. The left panels in Figure 8 show the response of two MOx sensors to the same binary mixtures (1:1 and 3:1 molar ratios of 1-hexene and 1-octene), as well as the expected results of linearly combining the power law response curves. The right panels show the same information, but for 3:1 and 1:10 mixtures of  $\alpha$ -pinene and 2-heptanone. For both sensors and all mixtures, the observed mixture response is far below this predicted sum, at least at higher mixing ratios (>10 ppb). This indicates that different VOCs can interact with each other on the MOx sensor, leading to a response that is not a linear combination of the individual power law response curves. This is consistent with the work of Llobet et al. (1998), who showed that linear addition was an acceptable approximation at low mixing ratios, but found that MOx responses at higher mixing ratios required the inclusion of an interaction term for each gas pair from the total sum of responses. Llobet et al. found that

the response of a MOx sensor to a binary mix of VOC<sub>1</sub> and VOC<sub>2</sub> could be represented as  $A_1[\text{VOC}_1]^{\beta_1} + A_2[\text{VOC}_2]^{\beta_2} - A_{12}[\text{VOC}_1]^{\beta_1} [\text{VOC}_2]^{\beta_2}$  (Llobet et al., 1998).

We applied this equation to our own mixture data, calculating a coefficient  $A_{12}$  that represents the interaction between the two species. For the 1-hexene/1-octene mixture, calculated values of  $A_{12}$  were found to be consistent across the different mixture proportions: for the MOx type 1 sensor, we found  $A_{12} = 2.9 \times 10^{-3} (\pm 1.3 \times 10^{-4})$  and  $A_{12} = 3.4 \times 10^{-3} (\pm 2.0 \times 10^{-4})$  for the 1:1 and 3:1 mixtures, respectively; for the MOx type 2 sensor,  $A_{12} = 5.3 \times 10^{-4} (\pm 1.3 \times 10^{-4})$  and  $A_{12} = 5.7 \times 10^{-4} (\pm 2.0 \times 10^{-4})$  for the 1:1 and 3:1 mixtures, respectively. Calculated  $A_{12}$  values for the  $\alpha$ -pinene and 2-heptanone mixture were also consistent: for the MOx type 1 sensor we found  $A_{12} = 3.6 \times 10^{-3} (\pm 6.0 \times 10^{-4})$  and  $A_{12} = 3.9 \times 10^{-3} (\pm 1.9 \times 10^{-4})$  for the 3:1 and 1:10 mixtures, respectively; for the MOx type 2 sensor,  $A_{12} = 1.3 \times 10^{-3} (\pm 3.3 \times 10^{-4})$  and  $A_{12} = 8.7 \times 10^{-4} (\pm 1.7 \times 10^{-4})$  for the 3:1 and 1:10 mixtures, respectively. However, we note the fits for the  $\alpha$ -pinene and 2-heptanone mixture (with  $R^2$  values ranging from 0.77 to 0.92) are poorer than those of the 1-hexene and 1-octene mixture ( $R^2$  ranging from 0.94 to 0.99). Overall, our results suggest that while the MOx signals may be additive at low levels of VOC (10 ppb or lower), the additivity approximation is inaccurate at higher levels, and that the inclusion of VOC-VOC interaction terms (Llobet et al., 1998) is necessary for accurate estimates of VOC levels.



**Figure 8:** Left panels: MOx responses to 1:1 (blue) and 3:1 (dark purple) mixtures of 1-hexene (red) and 1-octene (magenta). Right panels: MOx responses to 3:1 (blue) and 1:10 (teal) mixtures of  $\alpha$ -pinene (indigo) and 2-heptanone (aquamarine). Solid lines: best fit of each VOC or mixture; dotted lines: the predicted mixture signal, assuming additivity; dashed lines: predicted mixture signal that includes a fitted interaction term (Llobet et al., 1998). The mixing ratio (x axis) refers to the total amount of VOC (i.e., both components of the mixtures) introduced to the sensors.

*Lines 422-427: Sensor colocation with reference instruments and “novel application of data analysis techniques that directly interpret air sensor measurements” has already been done extensively with VOC sensors, which should be acknowledged. A few examples include: Hong, Gung-Hwa, et al. "Long-term field calibration of low-cost metal oxide VOC sensor: Meteorological and interference gas effects." Atmospheric Environment 310 (2023): 119955. , Frischmon, Caroline, et al. "Improving the quantification of peak concentrations for air quality sensors via data weighting." Atmospheric Measurement Techniques 18.13 (2025): 3147-3159. And Okorn, Kristen, and Michael Hannigan. "Applications and limitations of quantifying speciated and source-apportioned vocs with metal oxide sensors." Atmosphere 12.11 (2021): 1383.*

Response: We thank the reviewer for these examples from the literature and have edited the manuscript to reference this prior work. We note these studies examined total VOCs, or a small handful of individual VOCs, and not for many VOCs within a complex mixture. We have added the following text:

“In fact, previous co-location studies with individual or small arrays of MOx sensors have yielded calibrations that have moderate correlation ( $R^2 < 0.7$ ) with total VOC or a handful of key VOCs (e.g., benzene, toluene, ethylbenzene, and xylenes) (Frischmon et al., 2025; Hong et al., 2023; Okorn and Hannigan, 2021); comparisons of measurements from larger sensor arrays to speciated VOC measurements (e.g., from PTR-MS) could improve characterization of complex VOC mixtures still further.”

*Line 449: Discussion of how the “chemically specific information about VOCs” would actually work in practice is lacking. The fingerprints shown in Figure 4 are certainly interesting, but how would they be applied or used in contexts where there is a complex mixture of compounds (thereby making it difficult to distinguish the individual fingerprints).*

Response: Consistent with our response to the comment on Figure 4, here we now add that “these ‘fingerprints’ can potentially be used in conjunction with pattern recognition techniques to characterize complex mixtures of VOCs.”

## Reviewer 2

*This paper provides a valuable demonstration of how the characteristics of different OEM VOC sensors can be leveraged to build an array capable of identifying individual compounds. A measurement system such as this would be extremely useful in ambient and indoor air quality studies, thus this work demonstrating these fundamental characteristics in a controlled laboratory setting is particularly helpful. Furthermore, the in-depth look at the impact of RH (especially the complicated effects for MOx sensors) was appreciated.*

Response: We thank the reviewer for this evaluation of our manuscript and thoughtful suggestions on how to improve it.

*Comment 1: Was the variability of the sensor array's response to each compound examined? (i.e., how consistent are the "fingerprints" in Figure 4?) I'm not clear on whether the array was exposed to each compound through one or more calibration sequences. If yes, it would be good to share these results. If not, consider including this in the discussion of the limitations.*

Response: The responses in Figure 4 represent one calibration sequence per compound. We have edited the manuscript to include this limitation, specifically including the following text under Figure 4:

“Each subplot represents sensor responses to a single calibration sequence.”

*Comment 2: Why were 1-hexene and 1-octene selected as the only multi-compound mixture to expose the array to? A mixture of compounds producing very different "fingerprints" of response from the array would be interesting as well (e.g., 1-hexene or 1-octene vs acetone or isoprene). If additional mixtures were examined, this would be very useful data to add as well, or recommend discussing how this multi-compound mixture was selected.*

Response: We agree that a different mixture would provide more insight into sensor responses. We have edited the manuscript to include results from a binary mixture of *a*-pinene and 2-heptanone. For more detail, see our response to Reviewer 1 above.

### Minor Comments

- *Lines 11 - 14: this sentence is a little long, recommend breaking up key points for readability*
- *Line 19: "measuring environmental VOCs" - maybe specify 'speciated VOCs', or 'individual VOCs', or 'differentiating individual VOC compounds', etc.*
- *Lines 42 - 43: I don't think this sentence needs to be in parentheses, recommend removing the parentheses*
- *Line 98: might be more appropriate to say "has not been fully explored"*
- *Lines 85 - 98: some additional papers it may be helpful to review/consider including (to give a fuller picture of related work):*
  - *Leidinger et al., 2014 on the identification on individual VOCs (for indoor air quality) using a sensor array*
  - *Leidinger et. al., 2016 on the incorporation of a pre-concentrator to reach sub ppm levels of quantification for VOCs*
  - *Sauerwald et al., 2018 on the detection of benzene in the presence of interferents*
  - *Collier-Oxandale et. al., 2020 on the incorporation of non-VOC sensors to inform sources of VOCs*

Response: We thank the reviewer for these suggestions, and have edited the manuscript accordingly, including citing all the suggested references.

## References

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